Effects of acid deposition on soil: An overview

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Introduction

Large amounts of chemical compounds are deposited to terrestrial ecosystems as a result of the emission of pollutants to the atmosphere. The present paper is an overview of the effects of acid deposition on soils, and includes only those compounds having the potential to influence the acid/base properties of soils. The paper is thus restricted to a discussion of the effects of the deposition of H-ions and the accompanying anions SO_4^{2-} and NO_3^- , as well as compounds having the potential to produce acids in the soil immediately or later. This last mentioned group includes compounds like SO_2 , NO_x and also NH_4^+.

Soil acidity is the main factor to be considered when evaluating the effects of acid deposition on soil. We know that during soil formation all soils in areas with a surplus of precipitation tend to become more acid. Soil acidification is a natural process and many of the compounds being deposited from the polluted atmosphere occur naturally in soils. The problem of assessing their effects is therefore one of quantification.

In the present paper a comparison is made of the amount of acid deposited from polluted atmosphere and the amount produced in forest soil. By comparing these two amounts with the sinks of H-ions in the forest it is possible to indicate how the acid/base properties of forest soil are influenced in areas exposed to acid deposition. The paper also includes a review of studies aimed more directly at examining effects of acid deposition on soil-base properties.

Natural H-ion production

As illustrated in Figure 1, H-ions are produced in different parts and in different processes by the ecosystem. In unpolluted atmosphere H-ions enter the system with precipitation for example in the form of carbonic acid. H-ions may be taken up in the canopy, as is often the case with deciduous trees, or it may be released, often from conifers. In the soil CO_2 production by soil organisms and plant roots is also a source of carbonic acid. H-ions are also produced when N- and S-compounds are mineralized. Nutrient uptake by plants may produce H- or bicarbonate-ions depending on the

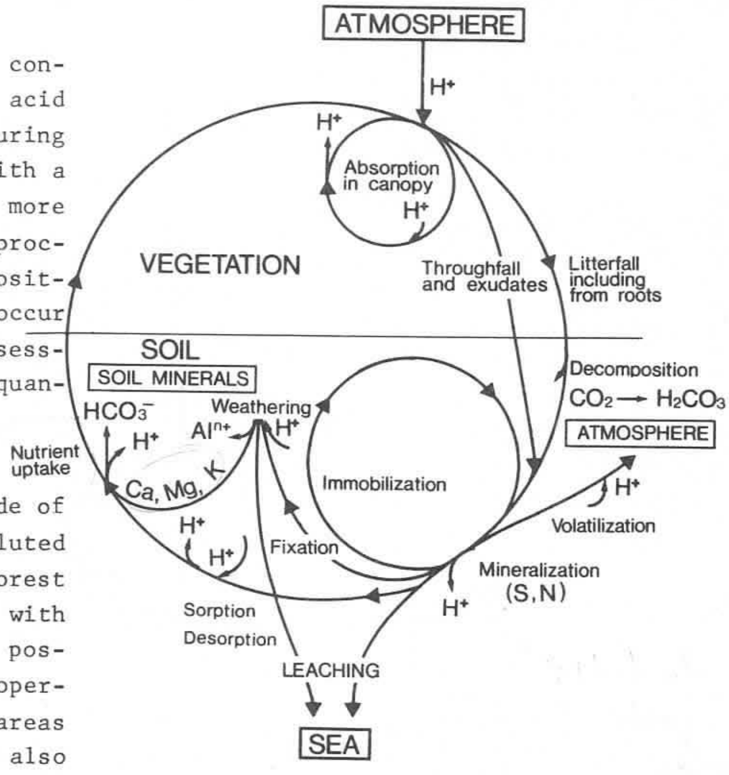

Figure 1. Simplified diagram of the nutrient cycling in terrestrial ecosystems.

Table 1. Amounts of H-ions produced in forest ecosystem.

Location	Site description	H^+ production me/m^2 yr.	Calculated as	Reference
Sweden	Pine, high productivity	300	Excess cation accumulation in trees and humus	1
	Pine, low productivity	∼ 50		
Hubbard Brook Experimental Forest U.S.A.	Deciduous forest	122	Total H^+ sources in the watershed minus the atmospheric input	2
Sweden	Pine, low productivity		Cation/anion balance	3
	Mineralization	223		
	Root uptake	309		
Solling, FRG	Beech	216	Cation/anion balance	4
	Spruce	346		

ratio of cation to anion uptake. Forest trees usually take up more cations than anions; therefore H-ion release is usually connected to the nutrient uptake in forest soil. When humus material accumulates in the soil H-ions also accumulate. Weathering processes are the main sink for H-ions, but H-ions may also be consumed by volatilization of S- and N-compounds.

The main part of the H-ion production in the soil can be estimated by the accumulation of humus and by the accumulation of excess cations in the biomass. As a forest grows there is an accumulation of base cations in the biomass and a corresponding accumulation of H-ions in the soil. However, provided that the organic material produced is decomposed completely to CO_2 , water and inorganic compounds, and that the biomass produced is not removed from the forest, there should be no long term net accumulation of H-ions. On the other hand, if the organic material is not completely decomposed and/or forest products are removed, the acidifying effect corresponds to the net uptake of cations over elements taken up as anions.

According to the factors mentioned above, the natural H-ion production in forest ecosystems will vary with soil properties and productivity, tree species, climate, forest management, etc. As shown by Nilsson et al. (1), it is also highly dependent on the age of the forest stand. After forest regeneration it increases up to a stand age of about 20 years, whereafter it gradually declines. Estimates of the total H-ion production as annual averages for an entire rotation period are given in Table 1. This shows that the average annual H-ion production appears to vary from about 50 me/m^2 yr in a poor pine forest to about 350 me/m^2 yr in a spruce forest in Germany.

The long-term acidifying effect of this H-ion production depends on harvesting policy. By increased removal of forest products the net acid production also increases. If only the stems including bark are harvested the net annual average H-ion production for the entire rotation period is of the magnitude 50 me/m^2 yr. If, however, whole trees are harvested both in thinnings and in the final clearing the figure is of the magnitude 120 me/m^2 yr (5, 6).

Variation in soil properties

The effects of acids on soil acid/base properties vary enormously with climatical conditions and soil properties. Leaching of plant nutrients only occurs in areas where precipitation exceeds evapotranspiration. Some of the more important soil properties are texture, amount of organic material, mineral composition, cation exchange capacity, base saturation, anion sorption capacity and soil depth. Variation in these properties is reflected on soil maps. Restricting the discussion to European conditions, the

FAO-Unesco soil map illustrates some of this variation (7). In Scandinavia and northern USSR the Orthic and Gelic Podzols dominate. Limited by low temperatures, the forests here are dominated by conifers and birch. At the other extreme, in the Mediterranean climatic belt extending from Portugal to Azerbaijan on the Caspian Sea, severe moisture stress is the dominant limitation. The soils are mainly Calcic and Eutric Cambisols, Chromic Luvisols and Xerosols. Between these extremes many other soil units are represented, ranging from the Calcaric Phaeozems, Calcic Chernozems and Gleyic Luvisols rich in bases to the thin Lithosols and eluviated and acid Podzols.

Effects of acids on soil

Cations exist in the soil in three phases: In the soil solution, adsorbed to the negatively charged soil particles, and in the minerals. Changes in the concentration in one of the phases influence the concentration in the others. Thus if the concentration of H-ions and an associated anion increase in the soil solution, H-ions will exchange with cations on the surface of the soil particles. Other cations may then move into the soil solution, and the H-ion saturation of the soil will increase. Increased acidity is again likely to increase the release of elements from the minerals by increased weathering rate.

Occurrence of cations in the soil solution is one premise for leaching. Another premise is the occurrence of a mobile anion. Anions like Cl^-, NO_3^-, HCO_3^- and often SO_4^{2-} are weakly sorbed to the soil particles. Therefore the presence of mobile anions is often considered to be the driving force in leaching (11). The anions of greatest interest in connection with acid deposition are NO_3^- and SO_4^{2-}. NO_3^- is weakly adsorbed to the soil particles, but in most natural soils it is readily taken up by plant roots and soil organisms. In this way NO_3^- is normally leached in small quantities in natural forest soils. Abrahamsen (12) compared the input of N by wet deposition with the runoff from forest watersheds and in lysimeters with forest soils in Europe and North America. On the average, the leaching of N amounted to only 30% of the incoming N by wet deposition. If including the dry deposition the relative leaching of N would be even lower. Presuming negligible denitrification, this means that more than 70% of the incoming N will be adsorbed in the soil/plant system. NO_3^- contributes therefore relatively little to the leaching of cations from the soils referred to, except during snowmelt, when the biological activity is low (13). It must also be mentioned that NO_3^- leaching appears to have increased in many countries with high atmospheric deposition such as Germany, the Netherlands, southern Sweden etc. This may indicate some kind of N saturation of the soil/plant systems.

Soil acidity increases and base saturation decreases when H-ions exchange with base cations adsorbed to the soil particles. The efficiency with which H-ions exchange with base cations is dependent on the base saturation, soil pH and the relative proportions of permanent and variable charges in the soil. A low base saturation, a low soil pH and a relatively high proportion of permanent charges will result in a lower replacing efficiency of H-ions (8, 9). In accordance with this, Bergseth (10) found a higher ratio of replaced amounts of Ca and Mg to sorbed H-ions in brown earth than in podzol.

SO_4^{2-} may in contrast to NO_3^- be sorbed in soils rich in amorphous oxides and hydroxides of Al and Fe. Such compounds are not as abundant in the soils of the sensitive areas in North America and Europe as in the soils developed for a longer time under warmer climates. The biological demand for S is much less than for N. The result of these factors is therefore that SO_4^{2-} appears to be weakly retained in soil/plant systems in the temperate zone. This is evident from the relationship between the wet deposition and leaching of SO_4^{2-} from forest watersheds or lysimeters. On the average slightly more S is leached than comes in by wet deposition. If including dry deposition, the conclusion is that the leaching of S is slightly less than the total deposition of S (12). Therefore SO_4^{2-} leaching is a crucial factor for the leaching of cations from soils.

The main sink of H-ions in soils is the weathering reactions. During weathering H-ions replace other elements in the minerals, and are therefore neutralized . Weathering rates are very difficult to estimate under natural conditions. For this reason our knowledge in this field is scanty. In Numedal in Norway it was found that the mean yearly weathering in podzols was equivalent to 20-140 me/m^2 yr (14). The long-term cationic denudation in the Hubbard Brook ecosystem was measured at 127 me/m^2 yr, but if the long-term accumulation of nutrients in the biomass supplied from the weathering reactions were included, the weathering rate would amount to about 200 me/m^2 yr (15). The H-ion consumption by weathering at four localities in Sweden was in the range of 35-79 me/m^2 yr (e.g. 1). In Glendye, Scotland, Reid et al. (16) calculated the loss of Mg^{2+}, Ca^{2+}, Na^+ and K^+ due to weathering to amount to 173 me/m^2 yr. The weathering rate is dependent on the climate, topography, parent material and biosphere; differences in these factors will give different weathering rates. Weathering rates also depend on the concentration of acid in the soil and are likely to increase with increased acid concentration.

It should also be mentioned that the effect of acids on soil depends on the pattern of infiltration and flow through the soil. This is mainly governed by the soil structure, the distribution of large and small pores and by the quantity of water flowing through the soil.

Effects of atmospheric acid deposition on soils

The information given in the preceding chapters tells us that the effect of the atmospheric deposition of acid or acid-forming substances will vary from one area to another depending on soil and vegetation properties. In addition comes the tremendous variation in deposition. The potential effect on soils is much higher in Central Europe, where the deposition of S amounts to 10 g/m^2 yr or even more, than in northern Scandinavia, where the deposition equals 0.5-1 g/m^2 yr or less. In this respect it is also important to remember that atmospheric pollutants in many areas of Central Europe have been deposited since the industrial revolution about 250 years ago. In Scandinavia the main increase in deposition came after World War II.

Research on the effects of acid deposition on soils during the last 10 years has shown that no single research strategy, e.g. experiments, modelling or field observations, can answer the question: What is the effect of a given atmospheric deposition on the acid/base properties of a given soil? The answer to this question must be based on the common multiple of the knowledge gained from all the research activities within this field. These activities are:

1. Use of general knowledge of soil chemistry - modelling.
2. Studies of soil chemistry as related to distance from large SO_2 emitters.
3. Acidification experiments.
4. Comparison of soil chemistry over time.

1. General knowledge of soil chemistry.

The discussion in the preceding chapters has demonstrated that in temperate forest soils:

- -Sulphate is commonly a mobile anion.
- -The efficiency of H-ions to exchange with base cations is commonly greater than 0.3.
- -The natural H-ion production in a growing forest ecosystem appears to be of the magnitude 50-350 me/m^2 as annual average for one rotation period.
- -The long-term net natural H-ion production (more rotation periods) in managed forests depends on the amount of the biomass removed. If only the stems (including

bark) are removed the H-ion production may be on the order of <10-50 me/m^2 yr. If whole trees are removed as much as 120 me H-ions/m^2 may be added to the system annually.

-The neutralizing capacity by weathering processes appears in podzolic soils to be on the order of 20-200 me/m^2 yr.

The conclusion from the above is that the natural H-ion production in managed forests may be of almost the same magnitude as the weathering rate. An additional input of acid would therefore tend to acidify soils unless the weathering rate is increased. However, most of the weathering rates given are from recent years and therefore more or less under the influence of acid deposition. In any case, studies on weathering rates are of great importance in solving this problem.

2. Effects of large SO_2 emitters.

The early literature on the effect of SO_2 on soil goes back to the beginning of this century. These studies are reviewed by Katz et al. (17), who also studied soil pH and base saturation at different distances from a large SO_2 emitter at Trail, British Columbia. The conclusion of this study was that pH and base saturation were depressed up to a distance of about 15 km from the source. A similar conclusion was reached from a study in the Sudbury area (18). In a later review Nyborg and Walker (19) concluded that the great majority of researchers showed evidence that SO_2 causes soil acidification in the vicinity of large sources, but the rate of the process was not well defined.

3. Experiments with artificial acidification.

Long-term effects of acid deposition may be simulated by applying water with a higher concentration of acids than in normal acid precipitation. Experiments of this kind include large-scale field plot- and lysimeter-studies as well as studies carried out on a laboratory scale. Laboratory and greenhouse studies have been carried out with the use of simulated rain of different acidities (20, 21, 22, 23). In the studies referred to, the simulated rain also contained normal concentrations of electrolytes like NO_3^-, NH_4^+, Cl^-, Na^+, K^+, Mg^{2+} and Ca^{2+}. All studies have shown increased leaching of base cations with increasing acidity of the simulated rain. Compared to the effect of control "rain" (pH 5.3-5.6) effects of "pH 4 rain" on the exchangeable base cation content of the soil were only slight. At pH 3, however, all studies have given similar results: Significant reductions in the content of exchangeable base cations in the soil. At this pH level relatively little damage is done to the ground vegetation, except for mosses which appear to be more sensitive to acid water than higher plants. At lower pH in "rain" more damage is done on the plants and part of the very significant reduction in the exchangeable cations is likely to be caused by this.

Results of acid application in field plots or field lysimeters show results similar to those mentioned for laboratory experiments. However, due to the size of the field plots and the huge amounts of water that therefore have to be applied, it has not been possible to obtain the normal concentration of electrolytes in the artificial rain in these studies. In Swedish experiments the acid applied was in the form of a quite strong solution of sulphuric acid and in amounts corresponding to a total of almost 200 kg S/ha over a 6-year period (24). This appears to have resulted in a statistically significant decrease in the content of "base cations" in all soil layers down to the C horizon except in the eluviated layer. In Norwegian experiments sulphuric acid was mixed with ground water and applied to field plots and lysimeters in concentrations corresponding to pH about 6 (control), pH 4, pH 3, and in some experiments also 2.5 and/or 2.0. The ground water has higher concentrations of, among others Ca^{2+} and Mg^{2+} than the precipitation. This has reduced the acidifying effect of the water applied compared to the effect of natural acid rain with similar pH levels. To overcome this

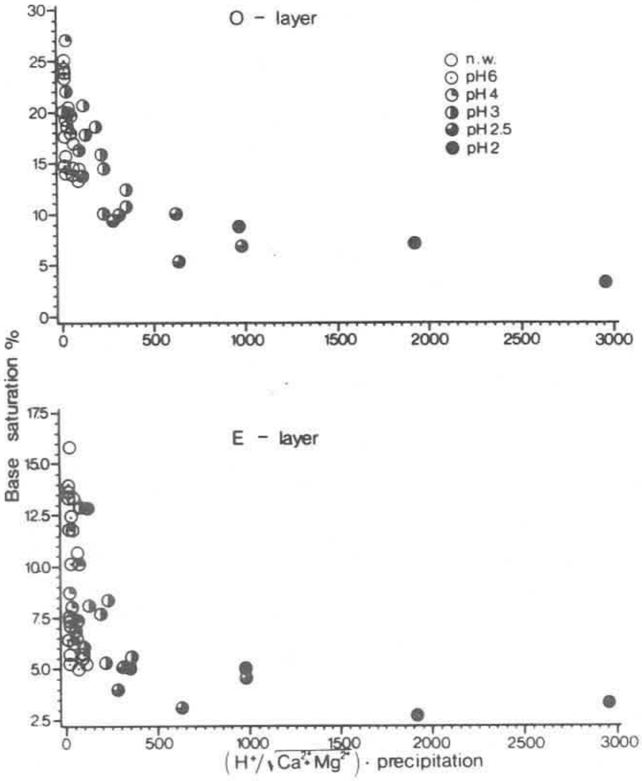

Figure 2. The relation between base saturation in the O- and E-layers and the total amount of acid applied to the field plots at the time of sampling. (See text for further explanation.)

problem the lime potential $(pH-1/2p(Ca^{2+}+Mg^{2+}))$ of the artificial rain should be used rather than the pH. The lime potential is actually the logarithm of the activity ratio $a_{H^+} / \sqrt{a_{Ca^{2+}+Mg^{2+}}}$ in solution. If multiplying the antilogarithm of the negative value of the lime potential of the artificial rain with the amount of water applied, we will get an expression for the total amount of acid applied to the plots. These figures can then be compared to corresponding figures for natural acid rain. An example may clarify this. Let us suppose we have two "rain" qualities A and B. These are:

A: pH 4, $(Ca^{2+}+Mg^{2+})$ 0.00001 M, LP = 1.5.

B: pH 4, $(Ca^{2+}+Mg^{2+})$ 0.0001 M, LP = 2.0.

This means that 1000 mm rain of quality A would be equal to 3162 mm rain of quality B. Figure 2 shows the relation between base saturation in the O-and the E-layers and the total amount of acid applied to the field plots at the time of sampling. The total amount of acid is calculated in the way described above. It is seen that there is a clear relationship between the amount of acid applied and the base saturation in the O-and E-layers of these podzol soils. In the Bs1- and Bs2-layers at approximately 10 and 25 cm soil depths respectively, no effect of the acid application could be observed. The largest decrease in base saturation is found in the "pH 2" treatment. Effects of this treatment cannot be compared directly to the long-term effect of natural acid precipitation with a pH of about 4.3. The reason is partly that large parts of the ground vegetation are killed by this treatment and partly that the equilibrium conditions in the soil will be quite different from those created by higher pH levels. The "pH 3" treatment, however, does not produce large damage to the ground vegetation except for some moss species. Therefore this treatment is more comparable to the effect of "naturally acid" rain. With these reservations in mind, the results of the experiments indicate that acid rain of pH 4.0-4.3 over some decades is likely to reduce the base saturation of the soils examined.

4. Soil resampling and reanalysis of acid/base properties.

Studies of changes over time in acid/base properties of the soil have been carried out in Sweden (26, 27), Canada (28) and in the FRG (29, 30). In one of the Swedish studies (26) no change in soil pH was found in an iron podzol during the period 1934 to 1973. The Canadian study included six soil types of which five were sandy and podzolized. In these five soil types no significant change in soil pH was found for the period 1960 to 1978. In the sixth soil type, a nonpodzolized silty clay loam, a large decrease in soil pH was found. However, in this case, the earliest measurement was questioned. The conclusion of the Canadian study was, therefore, that no significant change in soil pH was likely during the period 1960 to 1978. The other Swedish study is based on about 30,000 analyses of pH. Exchangeable cations were analysed for about 2,500 randomly selected test plots situated between 59° and 61° N (27). During the period 1961/63 to 1971/73 a slightly significant increase in exchangeable acidity was found.

At the same time a very significant decrease in the contents of exchangeable Ca, Mg and K was observed. The German studies are from a Berlin forest (29) and from different forest areas in North Rhine - Westphalia (30). In the forest of Berlin a very significant decrease in pH of the upper 10 cm of the soil was found during the period 1950 to 1980 for the three soil types "Rostbraunerden", "Vergleyte Rostbraunerden" and "Gleye". The results may also indicate decreased pH at 100 cm soil depth, but statistical analysis of these data showed no significant differences. In North Rhine-Westphalia 35 sites were sampled including soils from podzols, brown earth and base-rich gley soils. The soils studied were at different distances from the Ruhr area and with an average distance of 45-65 km. Almost all soils showed decreased soil pH during the period 1959/61 to 1981. The pH change was smallest for the most acid soils (pH 2.5-2.9) and largest for the least acid soils (pH 5.0 - 5.4). No relationship between the change in soil pH and distance from the Ruhr area was found.

The interpretation of the studies referred to is not straight-forward. Changes in the acid/base properties of the soil may be caused by various silvicultural measures such as fertilization, increased utilization of forest biomass, or increased stocking of the forest stands. However, as the most significant changes have occurred in Central Europe where the atmospheric depositions are largest, the soil changes appear at least partly to have been caused by these depositions.

Conclusions and summary

The acidifying effect of acid deposition on soil and water depends on the balance between the sum of the internal H-ion production and the atmospheric deposition, and the consumption of H-ions in weathering reactions. The acidifying effect on soil is also modified by the replacing efficiency of H-ions on the soil colloids and by the SO_4^{2-} sorption capacity of the soil. In temperate forest soils SO_4^{2-} sorption capacity is generally low and the replacing efficiency of H-ions appears mostly to be larger than 0.3-0.5. The internal H-ion production in a growing forest ecosystem appears to be of the magnitude 50 me/m^2 yr in poor pine forests to about 350 me/m^2 yr in fast-growing spruce forest. The net H-ion production of managed forests (more rotation periods) depends on the amount of biomass removed from the forest. If only the stems are removed the net H-ion supply to the ecosystem would be of the magnitude <50-10 me/m^2 yr. If whole trees are removed, the net production may be up to 120 me H /m^2yr. Estimates of weathering rates vary from 20 to 200 me/m^2 yr for podzolic soils. This means that in forests used for timber production there appears to be a near balance between the internal H-ion production and the consumption of H-ions by weathering. Because of the lack of studies on both internal H-ion production and weathering rate this conclusion must be considered as an hypothesis. However, if it proves to be correct, the atmospheric acid deposition will increase soil acidity. In this case it would also be possible to estimate the rate of acidification. The qualitative part of the hypothesis is supported by studies on soil acidity as dependent on distance from large SO_2 emitters, by studies with artificial acidification, and also by studies on changes in soil acidity over time in areas with different deposition rates.

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